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[Title of the Invention] WHITE ORGANIC ELECTROLUMINESCENCE

DEVICE

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[Title of the Invention] WHITE ORGANIC ELECTROLUMINESCENCE DEVICE

[Claims]

[Claim 1]

An organic electroluminescence device emitting white light which comprises a pair of electrodes, at least two light emitting layers and an electron transporting layer comprising a heterocyclic derivative having nitrogen atom or a heterocyclic derivative having silicon atom, the light emitting layers and the electron transporting layer being disposed between the pair of electrodes, wherein

an energy gap of a host compound comprised in each light emitting layer Eg(Host-i) satisfies following relation (I):

$$2.9 \text{ eV} \leq \text{Eg(Host-i)}$$
 .. (I)

(in the formula, Eg(Host-i) represents an energy gap of a host compound comprised in an i-th light emitting layer from the electron transporting layer, (i representing an integer of 1 to n),

an energy gap of the heterocyclic derivative having nitrogen atom or the heterocyclic derivative having silicon atom comprised in the electron transporting layer (Eg(ETM)) satisfies following relation (II):

$$2.9 \text{ eV} < \text{Eg}(\text{ETM})$$
 .. (II)

and

an ionization potential of a host compound comprised in a light emitting layer adjacent to the electron transporting layer (Ip(Host-1)) and an ionization potential of the heterocyclic derivative having nitrogen atom or the heterocyclic derivative having silicon atom comprised in the electron transporting layer (Ip(ETM)) satisfy following relation (III):

$$Ip(ETM) \leq Ip(Host-1)+0.3 eV .. (III)$$
[Claim 2]

An organic electroluminescence device emitting white light according to Claim 1, wherein the energy gap of a host compound comprised in each light emitting layer Eg(Host-i) and the energy gap of the heterocyclic derivative having nitrogen atom or the heterocyclic derivative having silicon atom comprised in the electron transporting layer (Eg(ETM)) satisfy following relation (IV):

$$2.9~\text{eV} < \text{Eg(ETM)} \leq \text{Eg(Host-i)} \dots \text{(IV)}$$
 [Claim 3]

An organic electroluminescence device emitting white light according to Claim 1 or 2, wherein at least one light emitting layer comprises a dopant having an energy gap of 2.9 eV or smaller.

#### [Claim 4]

An organic electroluminescence device emitting white light according to any one of Claims 1 to 3, which comprises

at least two light emitting layers having different peak wavelengths of light emission.

# [Claim 5]

An organic electroluminescence device emitting white light according to any one of Claims 1 to 4, wherein a difference between a greatest peak wavelength of light emission and a second greatest peak wavelength of light emission is 50 nm or greater.

## [Claim 6]

An organic electroluminescence device emitting white light according to any one of Claims 1 to 5, wherein the electron transporting layer comprises a heterocyclic derivative having nitrogen atom represented by following general formula (1):

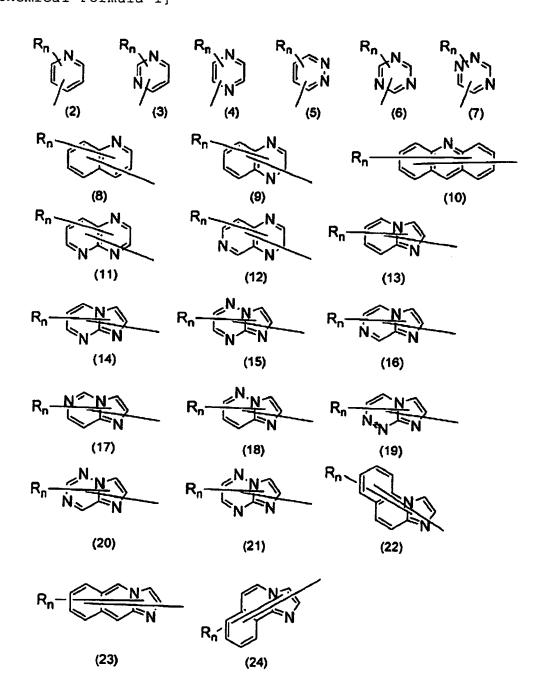
$$HAr-L-Ar^1-Ar^2$$
 (1)

(in the formula, HAr represents a substituted or unsubstituted heterocyclic group having nitrogen atom, Ar<sup>1</sup> represents a substituted or unsubstituted divalent aromatic hydrocarbon group having 6 to 40 carbon atoms, Ar<sup>2</sup> represents a substituted or unsubstituted aryl group having 6 to 40 carbon atoms or a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms, and L represents a single bond or a substituted or unsubstituted arylene group).

#### [Claim 7]

An organic electroluminescence device emitting white light according to Claim 6, wherein the HAr represents a

heterocyclic group having nitrogen atom represented by one of following formulae (2) to (24), and formula (A): [Chemical Formula 1]



(in the formula, R represents a substituted or unsubstituted aryl group having 6 to 40 carbon atoms, a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms or a substituted or unsubstituted alkoxyl group having 1 to 20 carbon atoms, n represents an integer of 0 to 5 and, when n represents an integer of 2 or greater, plural R may represent the same group or different groups, and the plural groups represented by R may be bonded to each other to form a cyclic structure);

[Chemical Formula 2]

$$\begin{array}{c|c} & & \\ \hline & & \\ Z & & \\ \hline \end{array} \begin{array}{c} & & \\ R' \\ \end{array} \begin{array}{c} & \\ X \end{array} \qquad (A)$$

(in the formula, plural  $R^1$  each independently represent a hydrogen atom, a halogen atom, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted aryl group having 6 to 40 carbon atoms, a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms or a group forming a condensed aromatic ring, Z represents an oxygen atom, a sulfur atom or a group represented by NR', (R' representing the same atom or group as that represented by  $R^1$ ), and x represents an integer of 2 to 8); or a substituted or unsubstituted carbazolyl group.

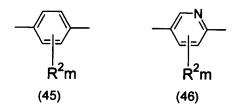
# [Claim 8]

An organic electroluminescence device emitting white light according to Claim 6, wherein the HAr represents a group expressed by one of following formulae (25) to (44): [Chemical Formula 3]

#### [Claim 9]

An organic electroluminescence device emitting white light according to any one of Claims 6 to 8, wherein the L represents a group represented by following formula (45) or (46):

### [Chemical Formula 4]



(in the formula,  $R^2$  represents a substituted or unsubstituted aryl group having 6 to 40 carbon atoms, a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms or a substituted or unsubstituted alkoxyl group having 1 to 20 carbon atoms, m represents an integer of 0 to 4 and, when m represents an integer of 2 or greater, plural  $R^2$  may represent the same group or different groups, and plural groups represented by  $R^2$  may be bonded to each other to form a cyclic structure).

#### [Claim 10]

An organic electroluminescence device emitting white light according to any one of Claims 6 to 9, wherein the  ${\rm Ar}^2$  represents a group represented by one of following formulae (47) to (53):

[Chemical Formula 5]

$$R^{3}p$$
 $R^{3}p$ 
 $R^{3}p$ 

(in the formula, R³ represents a substituted or unsubstituted aryl group having 6 to 40 carbon atoms, a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms or a substituted or unsubstituted alkoxyl group having 1 to 20 carbon atoms, p represents an integer of 0 to 9, q represents an integer of 0 to 5 and, when p or p+q represents an integer of 2 or greater, plural R³ may represent the same group or different groups, and plural groups represented by R³ may be bonded to each other to form a cyclic structure). [Claim 11]

An organic electroluminescence device emitting white light according to any one of Claims 6 to 10, wherein the  ${\rm Ar}^1$  represents a group represented by following formula (54) or

(55):

#### [Chemical Formula 6]

$$R^{5}$$
 $R^{6}$ 
 $R^{7}$ 
 $R^{8}$ 
 $R^{9}$ 
 $R^{10}$ 
 $R^{12}$ 
 $R^{17}$ 
 $R^{13}$ 
 $R^{16}$ 
 $R^{15}$ 
 $R^{15}$ 
 $R^{15}$ 
 $R^{15}$ 

(in the formula,  $R^4$  to  $R^{17}$  each independently represent a hydrogen atom, a halogen atom, a substituted or unsubstituted aryl group having 6 to 40 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 40 carbon atoms, a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms or a substituted or unsubstituted alkoxyl group having 1 to 20 carbon atoms, and  $Ar^3$  and  $Ar^4$  each represent a substituted or unsubstituted aryl group having 6 to 40 carbon atoms or a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms or a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms).

[Detailed Description of the Invention]

[0001]

[Technical Field to which the Invention Belongs]

The present invention relates to an organic electroluminescence device emitting white light, and more particularly to an organic electroluminescence device

emitting white light which provides a great efficiency of light emission under a low voltage, has a long lifetime and exhibits no change in the chromaticity.

[0002]

[Prior Art]

Organic electroluminescence (EL) devices which utilize organic substances are expected to be useful for application as inexpensive full color display devices of the solid light emission type having a great size, and various developments on the organic EL devices are being conducted. In general, an EL device has a construction comprising a light emitting layer and a pair of electrodes disposed at both sides of the light emitting layer. The light emission of the organic EL device is a phenomenon in which, when an electric field is applied between the two electrodes, electrons are injected from the cathode side and holes are injected from the anode side, the electrons are recombined with the holes in the light emitting layer to form an excited state, and energy generated when the excited state returns to the ground state is emitted as light.

Recently, organic EL devices emitting white light are attracting attention as the devices capable of the full color display using color filters in addition to applications as monocolor displays and lighting applications such as back light.

To produce the organic EL device emitting white light, a device having a light emitting layer of the laminate type which has plural light emitting layers is proposed (Patent Document 1). In the organic EL device having the light emitting layer of the laminate type, white light is emitted by doping each light emitting layer with a dopant, and light other than white light can also be emitted by changing the type of the dopant. However, the device having the light emitting layer of the laminate type has a problem in that the voltage necessary for the driving is great, the lifetime of light emission is short, and the chromaticity tends to change.

[Patent Document 1]

Japanese Patent Application Laid-Open No. 2002-093583 [0003]

[Problems that the Invention is to Solve]

The present invention has been made to overcome the above problem and has an object of providing an organic EL device emitting white light which provides a great efficiency of light emission under a low voltage, has a long lifetime and exhibits no change in the chromaticity.

[0004]

[Means for Solving the Problems]

As the result of intensive studies by the present inventors to achieve the above object, it was found that the above object could be achieved when the energy gap of a host

material comprised in a light emitting layer was in a specific range, the energy gap of a heterocyclic derivative having nitrogen atom or a heterocyclic derivative having silicon atom comprised in the electron transporting layer was in a specific range, and the ionization potential of the heterocyclic derivative having nitrogen atom or the heterocyclic derivative having silicon atom in the electron transporting layer and the ionization potential of the host material in the light emitting layer adjacent to the electron transporting layer satisfied a specific relation. The present invention has been completed based on the knowledge.

[0005]

That is, the present invention provides an organic EL device emitting white light which comprises a pair of electrodes, at least two light emitting layers and an electron transporting layer comprising a heterocyclic derivative having nitrogen atom or a heterocyclic derivative having silicon atom, the light emitting layers and the electron transporting layer being disposed between the pair of electrodes, wherein

an energy gap of a host compound comprised in each light emitting layer Eg(Host-i) satisfies the following relation (I):

 $2.9 \text{ eV} \leq \text{Eg(Host-i)} \qquad .. \quad (\text{I})$  (in the formula, Eg(Host-i) represents an energy gap of a host

compound comprised in an i-th light emitting layer from the electron transporting layer, (i representing an integer of 1 to n),

an energy gap of the heterocyclic derivative having nitrogen atom or the heterocyclic derivative having silicon atom comprised in the electron transporting layer (Eg(ETM)) satisfies the following relation (II):

$$2.9 \text{ eV} < \text{Eg(ETM)}$$
 .. (II)

and

an ionization potential of a host compound comprised in a light emitting layer adjacent to the electron transporting layer (Ip(Host-1)) and an ionization potential of the heterocyclic derivative having nitrogen atom or the heterocyclic derivative having silicon atom comprised in the electron transporting layer (Ip(ETM)) satisfy the following relation (III):

$$Ip(ETM) \le Ip(Host-1)+0.3 \text{ eV}$$
 .. (III)

[Mode for Carrying Out the Invention]

The construction of the layers in the organic EL device emitting white light of the present invention includes a constitution exhibited in Figure 1. Examples thereof are as follows:

(1) An anode/a light emitting layer/an electron transporting layer (/an electron injecting layer)/a cathode;

- (2) An anode/(a hole injecting layer/) a hole transporting layer/a light emitting layer/an electron transporting layer(/an electron injecting layer)/a cathode;
- (3) An anode/a layer emitting red light/a layer emitting blue light/an electron transporting layer(/an electron injecting layer)/a cathode;
- (4) An anode/(a hole injecting layer/) a hole transporting layer/a layer emitting green to red light/a layer emitting blue light/an electron transporting layer(/an electron injecting layer)/a cathode;
- (5) An anode/(a hole injecting layer/) a hole transporting layer/a layer emitting green to red light/a layer emitting white light/an electron transporting layer(/an electron injecting layer)/a cathode;
- (6) An anode/a layer emitting blue light/a layer emitting green to red light/an electron transporting layer(/an electron injecting layer)/a cathode;
- (7) An anode/(a hole injecting layer/) a hole transporting layer/a layer emitting blue light/a layer emitting green to red light/an electron transporting layer (/an electron injecting layer)/a cathode;
- (8) An anode/(a hole injecting layer/) a hole transporting layer/a layer emitting white light/a layer emitting green to red light/an electron transporting layer(/an electron injecting layer)/a cathode; and

(9) An anode/(a hole injecting layer/) a hole transporting layer/a layer emitting blue light/a layer emitting white light/an electron transporting layer(/an electron injecting layer)/a cathode,

and especially, device constructions (6) to (9) are preferable, and device construction (6) in which the layer emitting blue light is disposed at the side of the anode and the layer emitting green to red light is disposed at the side of the cathode is more preferable. Incidentally, the layer emitting white light means a light emitting layer emitting white light or light in the white area in the CIE chromaticity coordinates.

Incidentally, the layer emitting white light means a light emitting layer emitting light having a great half width (80 nm or greater) or light in the white area in the CIE chromaticity coordinates. The definition of the white light is defined by means of the CIE chromaticity coordinates in the diagram shown in Figure 2. Specific examples of the layer emitting white light include light emitting layers of the laminate type such as "a light emitting layer emitting blue light/a light emitting layer emitting green to red light" and "a light emitting layer emitting green to red light light emitting layer emitting blue light" described above, and light emitting layers using a light emitting material or a dopant exhibiting a great half width of light emission such as those described in Japanese Patent Application Laid-Open No. Heisei

8(1996) - 315983.

[0007]

In the organic EL device of the present invention having the above device construction, the energy gap of a host compound comprised in each light emitting layer Eg(Host-i) satisfies the following relation (I):

2.9 eV 
$$\leq$$
 Eg(Host-i) .. (I)

(in the formula, Eg(Host-i) represents an energy gap of a host compound comprised in an i-th light emitting layer from the electron transporting layer, (i representing an integer of 1 to n)), the energy gap of the heterocyclic derivative having nitrogen atom or the heterocyclic derivative having silicon atom comprised in the electron transporting layer (Eg(ETM)) satisfies the following relation (II):

$$2.9 \text{ eV} < \text{Eg}(\text{ETM})$$
 .. (II)

and the ionization potential of the host compound comprised in the light emitting layer adjacent to the electron transporting layer (Ip(Host-1)) and the ionization potential of the heterocyclic derivative having nitrogen atom or the heterocyclic derivative having silicon atom comprised in the electron transporting layer (Ip(ETM)) satisfy the following relation (III):

$$Ip(ETM) \leq Ip(Host-1)+0.3 \text{ eV}$$
 .. (III)

It is preferable that the following relations are satisfied:

Eg(Host-i)<3.2 eV
Eg(ETM)<3.2 eV
[0008]

The above energy gap can be obtained by preparing a dilute solution (a concentration of about  $10^{-5}\,\mathrm{M}$ ) of the object organic substance and calculating the energy gap based on the end portion of the absorption spectrum of the prepared solution at the side of the long wavelength.

The above ionization potential can be measured by, for example, a method in which the object organic substance is irradiated with light from a heavy hydrogen lamp via a monochromator for spectral diffraction, the discharge of photoelectrons is measured with an electrometer to obtain a curve showing the discharge of photoelectrons with the photon energy of irradiation, and obtaining the threshold value of the discharge of photoelectrons from the curve in accordance with the extrapolation method.

[0009]

In the organic EL device of the present invention, the host compound in the light emitting layer satisfies the above relation (I), the heterocyclic derivative having nitrogen atom or the heterocyclic derivative having silicon atom in the electron transporting layer satisfies the above relation (II), and the ionization potential of the host compound comprised in the light emitting layer adjacent to the electron

transporting layer and the ionization potential of the heterocyclic derivative having nitrogen atom or the heterocyclic derivative having silicon atom comprised in the electron transporting layer satisfy the above relation (III). Due to this condition, the hole barrier is optimized and is not excessively great or excessively small. No decrease in the hole injecting property or the electron injecting property takes place, and the holes and the electrons are very efficiently recombined in the light emitting layer. Thus, the organic EL device of the present invention provides a great efficiency of light emission even under a low voltage, has a long lifetime and exhibits no change in the chromaticity.

It is preferable that relation (III) is expressed as:

 $Ip(ETM) \le Ip(Host-1)+0.2 eV$ 

and more preferably as:

 $Ip(ETM) \le Ip(Host-1)+0.1 \text{ eV}.$ [0010]

In the organic EL device of the present invention, it is preferable that the energy gap of the host compound comprised in each light emitting layer Eg(Host-i) and the energy gap of the heterocyclic derivative having nitrogen atom or the heterocyclic derivative having silicon atom comprised in the electron transporting layer (Eg(ETM)) satisfy the following relation (IV):

2.9 eV 
$$<$$
 Eg(ETM)  $\le$  Eg(Host-i).. (IV)

while the above relations (I) to (III) are satisfied. [0011]

In the organic EL device of the present invention, it is preferable that at least one light emitting layer comprises a dopant having an energy gap Eg(Dop-i), (i representing an integer of 1 to n), of 2.9 eV or smaller. It is more preferable that the energy gap Eg of the light emitting layer forming the interface with the electron transporting layer is smaller than 2.65 eV, or at least one layer among the light emitting layers not forming the interface with the electron transporting layer, i.e., having a value of i of 2 or greater, comprises a dopant having the energy gap satisfying the following relation (V):

 $2.65~{\rm eV}~<~{\rm Eg(Dop-i)}~\leq~2.9~{\rm eV}~..~({\rm V})$  The device can emit light with stability when the above conditions are satisfied.

The concentration of the dopant in the light emitting layer is not particularly limited. The concentration of the dopant is preferably 0.05 to 15% by weight and more preferably 0.1 to 10% by weight. A single light emitting layer may be doped with plural types of dopant.

The device emitting white light can be obtained by the combination of light emitting layers of two or three colors. It is necessary for this construction that at least one dopant is added. When the energy gap of the dopant in at least one layer among the light emitting layers not forming the interface

with the electron transporting layer, i.e., having the value of i of 2 or greater, satisfies the above relation (V), the change in the color tone due to the absorption of the emitted light with the dopant and the re-emission of light thereafter taken place upon taking up the light at the anode side can be decreased.

It is not necessary that a single dopant is used in each light emitting layer in the present invention, and plural dopants can be used if needed so that the efficiency can be increased and the color tone can be adjusted. Examples of the form of doping include (1) partial doping of the same host compound, (2) combinations of different host compounds and different dopants and combinations of (1) and (2).

It is preferable that the organic EL device of the present invention comprises two or more light emitting layers having different peak wavelengths of light emission. It is more preferable that each light emitting layer comprises the above dopant. It is still more preferable in the organic EL device of the present invention that the difference between the peak wavelength of the greatest light emission and the peak wavelength of the second greatest light emission is 50 nm or greater.

[0012]

To achieve the stable light emission and increase the efficiency of light emission of the organic EL device, plural

electron transporting layers may be disposed, or a layer of an inorganic compound may be disposed between the anode and the light emitting layer or between the cathode and the electron transporting layer. Examples of the inorganic compound used in the layer of an inorganic compound include various oxides, nitrides, oxide nitrides and halides such as oxides of alkali metals, oxides of alkaline earth metals, oxides of rare earth metals, halides of alkali metals, halides of alkali metals, halides of rare earth metals, halides of rare earth metals, SiO<sub>x</sub>, AlO<sub>x</sub>, SiN<sub>x</sub>, SiON, AlON, GeO<sub>x</sub>, LiO<sub>x</sub>, LiON, TiO<sub>x</sub>, TiON, TaO<sub>x</sub>, TaON, TaN<sub>x</sub>, C, LiF, MgF<sub>2</sub>, CaF<sub>2</sub>, MgF<sub>2</sub> and NaF.

As the layer disposed between the anode and the light emitting layer, layers comprising  $SiO_x$ ,  $AlO_x$ ,  $SiN_x$ , SiON, AlON,  $GeO_x$  or C are preferable. As the layer disposed between the cathode and the electron transporting layer, layers comprising LiF, MgF<sub>2</sub>, CaF<sub>2</sub>, MgF<sub>2</sub> or NaF are preferable.

An oxidizing agent may be added into the organic layer adjacent to the anode. As the oxidizing agent, electron-attracting or electron-accepting oxidizing agents are preferable. Examples of such oxidizing agents include Lewis acids, various quinone derivatives, dicyanoquino-dimethane derivatives and salts formed with aromatic amines and Lewis acids. Examples of the Lewis acid include iron chlorides, antimony chloride and aluminum chloride. A reducing agent may be added to the organic layer adjacent to

the cathode. Examples of the reducing agent include alkali metals, alkaline earth metals, oxides of alkali metals, oxides of alkaline earth metals, oxides of rare earth metals, halides of alkali metals, halides of alkaline earth metals and halides of rare earth metals. Among the alkali metals, Cs, Li, Na and K are preferable.

[0013]

In the present invention, the light emitting material (the host material) which can be used for the light emitting layer is not particularly limited, and any material can be used as long as the material satisfies the above relation (I). Specific examples thereof include commonly used materials emitting blue light. The light emitting property of the light emitting material is not particularly limited. Any of the known fluorescent materials and phosphorescent materials may be used.

Specific examples of the material emitting blue light include anthracene derivatives, styryl derivatives, aromatic amines, aluminum chelates having mixed ligands and carbazole derivatives.

As the anthracene derivative, compounds having a phenyl-anthracene skeleton structure are preferable.

As the styryl derivative, distyryl derivatives, tristyryl derivatives, tetrastyryl derivatives and styrylamine derivatives are preferable.

As the aromatic amine, compounds having 2 to 4 nitrogen atoms substituted with an aromatic group are preferable, and compounds having 2 to 4 nitrogen atoms substituted with an aromatic group and having at least one alkenyl group are more preferable.

[0014]

Examples of the aluminum chelates having mixed ligands include  $bis(R^s-8-quinolinolato)$  (phenolato)aluminum(III) chelates represented by the following formula [viii]:

$$(R^S-Q)_2-Al-O-L$$
 [viii]

In the above formula, Q represents a substituted 8-quinolinolato ligand, R<sup>S</sup> represents a substituent to the 8-quinolinolato ring, O-L represents a phenolato ligand, and L represents a hydrocarbon group having an aryl portion.

As the aluminum chelate having mixed ligands, the following compounds are preferable:

[Chemical Formula 7]

Eq(ETM) = 3.0eV

Eg(ETM) = 3.0eV

As the carbazole derivative, the following compound, etc. are preferable:

# [Chemical Formula 8]

[0015]

Examples of the anthracene derivative include compounds represented by the following general formulae [i] to [v]: [Chemical Formula 9]

$$R^{9'}$$
 $R^{2'}$ 
 $R^{7'}$ 
 $R^{7'}$ 
 $R^{10'}$ 
 $R^{3'}$ 
 $R^{6'}$ 
 $R^{6'}$ 
 $R^{5'}$ 

[0016]

[in the formula, R¹' to R¹0' each independently represent a hydrogen atom, a halogen atom, a cyano group, a nitro group, a substituted or unsubstituted alkyl group having 1 to 20 carbon atom, a substituted or unsubstituted alkoxyl group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group

having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heteroaryl group having 5 to 30 carbon atoms.

 $\mathrm{Ar}^{1'}$  and  $\mathrm{Ar}^{2'}$  each independently represent a substituted or unsubstituted aryl group having 6 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 1 to 20 carbon atoms. The substituent includes substituted or unsubstituted alkyl groups having 1 to 20 carbon atom, substituted or unsubstituted alkoxyl groups having 1 to 20 carbon atoms, substituted or unsubstituted aryloxy groups having 6 to 30 carbon atoms, substituted or unsubstituted alkylthio groups having 1 to 20 carbon atoms, substituted or unsubstituted arylthio groups having 6 to 30 carbon atoms, substituted or unsubstituted arylalkyl groups having 7 to 30 carbon atoms, unsubstituted monocyclic groups having 5 to 30 carbon atoms, substituted or unsubstituted condensed polycyclic groups having 10 to 30 carbon atoms and substituted or unsubstituted heteroaryl groups having 5 to 30 carbon atoms].

[0017]

[Chemical Formula 10]

$$R^{9'} R^{2'} \longrightarrow R^{7'} R^{10'}$$
 $R^{3'} \longrightarrow R^{6'} \longrightarrow R^{6'}$ 
 $R^{4'} R^{5'}$ 

[0018]

[in the formula,  $R^{1'}$  to  $R^{10'}$  are as defined above.

 $\mathrm{Ar}^{3'}$  and  $\mathrm{Ar}^{4'}$  each independently represent a substituted or unsubstituted aryl group having 6 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 1 to 20 The substituent includes substituted or carbon atoms. unsubstituted alkyl groups having 1 to 20 carbon atom, substituted or unsubstituted alkoxyl groups having 1 to 20 carbon atoms, substituted or unsubstituted aryloxy groups having 6 to 30 carbon atoms, substituted or unsubstituted alkylthio groups having 1 to 20 carbon atoms, substituted or unsubstituted arylthio groups having 6 to 30 carbon atoms, substituted or unsubstituted arylalkyl groups having 7 to 30 carbon atoms, unsubstituted monocyclic groups having 5 to 30 carbon atoms, substituted or unsubstituted condensed polycyclic groups having 10 to 30 carbon atoms, substituted or unsubstituted heteroaryl groups having 5 to 30 carbon atoms or substituted or unsubstituted alkenyl groups having 4 to 40 carbon atoms. n' represents a number of 1 to 3, m' represents

a number of 1 to 3, and  $n'+m' \ge 2$ ].
[0019]

[Chemical Formula 11]

[in the formula,  $R^{1'}$  to  $R^{8'}$ ,  $Ar^{3'}$  and  $Ar^{4'}$  are as defined above] [0020]

[Chemical Formula 12]

$$(R^{1})_{a} \xrightarrow{R^{5}} L^{1} \xrightarrow{R^{9}} (R^{2})_{b}$$
 [iv]

[in the formula, R<sup>1"</sup> to R<sup>10"</sup> each independently represent a hydrogen atom, a substituted or unsubstituted alkenyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted cycloalkyl group having 3 to 20 carbon atoms, a substituted or unsubstituted aryl group having 6 to 30 carbon atoms, a substituted or unsubstituted alkoxyl group having 1

to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylamino group having 1 to 20 carbon atoms, a substituted arylamino group having 1 to 40 carbon atoms or a substituted or unsubstituted heteroaryl group having 5 to 30 carbon atoms, and a and beach represent an integer of 1 to 5. When a or b represents an integer of 2 or greater, atoms or groups represented by plural  $R^{1"}$  or plural  $R^{2"}$ , respectively, may be the same with or different from each other, and groups represented by plural  $R^{1"}$  or plural  $R^{2"}$ , respectively, may bond to each other to form a ring. A pair of groups represented by  $R^{3"}$  and  $R^{4"}$ ,  $R^{5"}$  and  $R^{6"}$ ,  $R^{7"}$  and  $R^{8"}$  or  $R^{9"}$  and  $R^{10"}$  may bond to each other to form a ring.  $L^1$  represents a single bond,  $-O^-$ ,  $-S^-$ ,  $-N(R)^-$ , (R representing an alkyl group or an aryl group which may be substituted), or an arylene group].

[0021]

# [Chemical Formula 13]

$$R^{13"}$$
 $R^{15"}$ 
 $R^{15"}$ 
 $R^{20"}$ 
 $R^{18"}$ 
 $R^{19"}$ 
 $R^{19"}$ 

[In the formula,  $R^{11}$ " to  $R^{20}$ " each independently represent an

atom or a group which is the same as that represented by  $R^{1"}$  to  $R^{10"}$ ; and c, d, e and f each represent an integer of 1 to 5. When c, d, e or f represents an integer of 2 or greater, atoms or groups represented by plural  $R^{11"}$ , plural  $R^{12"}$ , plural  $R^{16"}$  or plural  $R^{17"}$  respectively, may be the same with or different from each other, and groups represented by plural  $R^{11"}$ , plural  $R^{12"}$ , plural  $R^{16"}$  or plural  $R^{17"}$ , respectively, may bond to each other to form a ring. A pair of groups represented by  $R^{13"}$  and  $R^{14"}$  or  $R^{18"}$  and  $R^{19"}$  may bond to each other to form a ring. L<sup>2</sup> represents an atom or a group which is the same as that represented by  $L^1$ ].

[0022]

Specific examples of the groups represented by the above general formulae (i) to (v) include the following groups. The alkyl group include, for example, methyl group, ethyl group, propyl group, isopropyl group, n-butyl group, s-butyl group, isobutyl group, t-butyl group, n-pentyl group, n-hexyl group, n-heptyl group, n-octyl group, hydroxymethyl group, 1-hydroxyethyl group, 2-hydroxyethyl group, 2-hydroxyisobutyl group, 1,2-dihydroxyethyl 1,3-dihydroxy-isopropyl group, 2,3-dihydroxy-t-butyl group, 1,2,3-trihydroxypropyl group, chloromethyl group, 1-chloroethyl group, 2-chloroethyl group, 2-chloroisobutyl group, 1,2-dichloroethyl group, 1,3-dichloroisopropyl group, 2,3-dichloro-t-butyl group, 1,2,3-trichloropropyl group,

bromomethyl group, 1-bromoethyl group, 2-bromoethyl group, 2-bromoisobutyl group, 1,2-dibromoethyl group, 1,3-dibromoisopropyl group, 2,3-dibromo-t-butyl group, 1,2,3-tribromopropyl group, iodomethyl group, 1-iodoethyl group, 2-iodoethyl group, 2-iodoisobutyl group, 1,2-diiodoethyl group, 1,3-diiodoisopropyl group, 2,3-diiodo-t-butyl group, 1,2,3-triiodopropyl group, aminomethyl group, 1-aminoethyl group, 2-aminoethyl group, 2-aminoisobutyl group, 1,2-diaminoethyl group, 1,3-diaminoisopropyl group, 2,3-diamino-t-butyl group, 1,2,3-triamino-propyl group, cyanomethyl group, 1-cyanoethyl group, 2-cyanoethyl group, 2-cyanoisobutyl group, 1,2-dicyanoethyl group, 1,3-dicyanoisopropyl group, 2,3-dicyano-t-butyl group, 1,2,3- tricyanopropyl group, nitromethyl group, 1-nitroethyl group, 2-nitroethyl group, 2-nitroisobutyl group, 1,2-dinitroethyl group, 1,3-dinitroisopropyl group, 2,3-dinitro-t-butyl group and 1,2,3-trinitropropyl group.

[0023]

The alkoxyl group is a group represented by -OY, and examples of the group represented by Y include the groups described as the examples of the alkyl group.

The alkylthio group is a group represented by -SY, and examples of the group represented by Y include the groups described as the examples of the alkyl group.

[0024]

The aryl group includes, for example, phenyl group, 1-naphthyl group, 2-naphthyl group, 1-anthryl group, 2-anthryl group, 9-anthryl group, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, 1-naphthacenyl group, 2-naphthacenyl group, 9-naphthacenyl group, 1-pyrenyl group, 2-pyrenyl group, 4-pyrenyl group, 2-biphenylyl group, 3-biphenylyl group, 4-biphenylyl group, p-terphenyl-4-yl group, p-terphenyl-3-yl group, p-terphenyl-2-yl group, m-terphenyl-4-yl group, m-terphenyl-3-yl group, m-terphenyl-2-yl group, o-tolyl group, m-tolyl group, p-tolyl group, p-t-butylphenyl group, p-(2-phenylpropyl)phenyl group, 3-methyl-2-naphthyl group, 4-methyl-1-naphthyl group, 4-methyl-1-anthryl 4'-methylbiphenylyl group and 4''-t-butyl-p-terphenyl-4-yl group.

[0025]

The aryloxy group is a group represented by -OZ', and examples of the group represented by Z' include the groups described as the examples of the aryl group.

The arylthio group is a group represented by -SZ', and examples of the group represented by Z' include the groups described as the examples of the aryl group.

[0026]

The arylalkyl group includes benzyl group, 1-phenylethyl

group, 2-phenylethyl group, 1-phenylisopropyl group, 2-phenylisopropyl group, phenyl-t-butyl group,  $\alpha$ -naphthylmethyl group,  $1-\alpha$ -naphthylethyl group,  $2-\alpha$ -naphthylethyl group,  $1-\alpha$ -naphthylisopropyl group,  $2-\alpha$ -naphthylisopropyl group, β-naphthylmethyl group,  $1-\beta$ -naphthylethyl group,  $2-\beta$ -naphthylethyl group,  $1-\beta$ -naphthylisopropyl group,  $2-\beta$ -naphthylisopropyl group, 1-pyrrolylmethyl group, 2-(1-pyrroly1)ethyl group, p-methylbenzyl group, m-methylbenzyl group, o-methylbenzyl p-chlorobenzyl group, group, m-chlorobenzyl group, o-chlorobenzyl group, p-bromobenzyl group, m-bromobenzyl group, o-bromobenzyl group, p-iodobenzyl group, m-iodobenzyl group, o-iodobenzyl group, p-hydroxybenzyl m-hydroxybenzyl group, o-hydroxybenzyl group, p-aminobenzyl group, m-aminobenzyl group, o-aminobenzyl group, p-nitrobenzyl group, m-nitrobenzyl group, o-nitrobenzyl group, p-cyanobenzyl group, m-cyanobenzyl group, o-cyanobenzyl group, 1-hydroxy-2-phenylisopropyl group, 1-chloro-2-phenylisopropyl group, and the like.

[0027]

The monocyclic group includes groups derived from cyclopentane, cyclohexane, cycloheptane, and the like.

The condensed polycyclic group includes, for example, groups derived from anthracene, naphthalene, phenanthrene, pyrene, tetracene, coronene, chrysene, fluoresceine and

perylene.

[0028]

The heteroaryl group includes 1-azaindolidin-2-yl group, 1-azaindolidin-3-yl 1-azaindolidin-5-yl group, group, 1-azaindolidin-6-yl group, 1-azaindolidin-7-yl group, 1-azaindolidin-8-yl group, 2-azaindolidin-1-yl group, 2-azaindolidin-3-yl group, 2-azaindolidin-5-yl group, 2-azaindolidin-6-yl 2-azaindolidin-7-yl group, group, 2-azaindolidin-8-yl 6-azaindolidin-1-yl group, group, 6-azaindolidin-2-yl group, 6-azaindolidin-3-yl group, 6-azaindolidin-5-vl group, 6-azaindolidin-7-yl group, 6-azaindolidin-8-yl 7-azaindolidin-1-yl group, group, 7-azaindolidin-2-yl 7-azaindolidin-3-yl group, group, 7-azaindolidin-5-yl group, 7-azaindolidin-6-yl group, 7-azaindolidin-7-yl 7-azaindolidin-8-yl group, group, 8-azaindolidin-1-yl 8-azaindolidin-2-yl group, group, 8-azaindolidin-3-yl 8-azaindolidin-5-yl group, group, 8-azaindolidin-6-yl 8-azaindolidin-7-yl group, group, 1-indolidinyl group, 2-indolidinyl group, 3-indolidinyl group, 5-indolidinyl group, 6-indolidinyl group, 7-indolidinyl group, 8-indolidinyl group, 1-pyrrolyl group, 2-pyrrolyl group, 3-pyrrolyl group, pyradinyl group, 2-pyridinyl group, 3-pyridinyl group, 4-pyridinyl group, 1-indolyl 2-indolyl group, 3-indolyl group, 4-indolyl group, 5-indolyl group, 6-indolyl group, 7-indolyl group, 1-isoindolyl group,

2-isoindolyl group, 3-isoindolyl group, 4-isoindolyl group, 5-isoindolyl group, 6-isoindolyl group, 7-isoindolyl group, 2-furyl group, 3-furyl group, 2-benzofuranyl group, 3-benzofuranyl group, 4-benzofuranyl group, 5-benzofuranyl group, 6-benzofuranyl group, 7-benzofuranyl group, 1-isobenzofuranyl group, 3-isobenzofuranyl group, 4-isobenzofuranyl group, 5-isobenzofuranyl group, 6-isobenzofuranyl group, 7-isobenzofuranyl group, 2-quinolyl group, 3-quinolyl group, 4-quinolyl group, 5-quinolyl group, 6-quinolyl group, 7-quinolyl group, 8-quinolyl group, 1-isoquinolyl group, 3-isoquinolyl group, 4-isoquinolyl group, 5-isoquinolyl group, 6-isoquinolyl group, 7-isoquinolyl group, 8-isoquinolyl group, 2-quinoxalinyl group, 5-quinoxalinyl group, 6-quinoxalinyl group, 1-carbazolyl group, 2-carbazolyl group, 3-carbazolyl group, 4-carbazolyl group, 9-carbazolyl group, 1-phenanthridinyl group, 2-phenanthridinyl group, 3-phenanthridinyl group, 4-phenanthridinyl group, 6-phenanthridinyl group, 7-phenanthridinyl group, 8-phenanthridinyl group, 9-phenanthridinyl group, 10-phenanthridinyl group, 1-acridinyl group, 2-acridinyl group, 3-acridinyl group, 4-acridinyl group, 9-acridinyl group, 1,7-phenanthrolin-2-yl group, 1,7-phenanthrolin-3-yl group, 1,7-phenanthrolin-4-yl group, 1,7-phenanthrolin-5-yl group, 1,7-phenanthrolin-6-yl group, 1,7-phenanthrolin-8-yl group, 1,7-phenanthrolin-9-yl group, 1,7-phenanthrolin-10-yl

group, 1,8-phenanthrolin-2-yl group, 1,8-phenanthrolin-3-yl group, 1,8-phenanthrolin-4-yl group, 1,8-phenanthrolin-5-yl group, 1,8-phenanthrolin-6-yl group, 1,8-phenanthrolin-7-yl group, 1,8-phenanthrolin-9-yl group, 1,8-phenanthrolin-10-yl group, 1,9-phenanthrolin-2-yl group, 1,9-phenanthrolin-3-yl group, 1,9-phenanthrolin-4-yl group, 1,9-phenanthrolin-5-yl group, 1,9-phenanthrolin-6-yl group, 1,9-phenanthrolin-7-yl group, 1,9-phenanthrolin-8-yl group, 1,9-phenanthrolin-10-yl group, 1,10-phenanthrolin-2-yl group, 1,10-phenanthrolin-3-yl group, 1,10-phenanthrolin-4-yl group, 1,10-phenanthrolin-5-yl group, 2,9-phenanthrolin-1-yl group, 2,9-phenanthrolin-3-yl group, 2,9-phenanthrolin-4-yl group, 2,9-phenanthrolin-5-yl group, 2,9-phenanthrolin-6-yl group, 2,9-phenanthrolin-7-yl group, 2,9-phenanthrolin-8-yl group, 2,9-phenanthrolin-10-yl group, 2,8-phenanthrolin-1-yl group, 2,8-phenanthrolin-3-yl group, 2,8-phenanthrolin-4-yl group, 2,8-phenanthrolin-5-yl group, 2,8-phenanthrolin-6-yl group, 2,8-phenanthrolin-7-yl group, 2,8-phenanthrolin-9-yl group, 2,8-phenanthrolin-10-yl group, 2,7-phenanthrolin-1-yl group, 2,7-phenanthrolin-3-yl group, 2,7-phenanthrolin-4-yl group, 2,7-phenanthrolin-5-yl group, 2,7-phenanthrolin-6-yl group, 2,7-phenanthrolin-8-yl group, 2,7-phenanthrolin-9-yl group, 2,7-phenanthrolin-10-yl group, 1-phenazinyl 2-phenazinyl group, 1-phenothiazinyl group, 2-phenothiazinyl group, 3-phenothiazinyl group, 4-phenothiazinyl

10-phenothiazinyl group, 1-phenoxazinyl group, 2-phenoxazinyl group, 3-phenoxazinyl group, 4-phenoxazinyl group, 10-phenoxazinyl group, 2-oxazolyl group, 4-oxazolyl group, 5-oxazolyl group, 2-oxadiazolyl group, 5-oxadiazolyl group, 3-furazanyl group, 2-thienyl group, 3-thienyl group, 2-methylpyrrol-1-yl group, 2-methylpyrrol-3-yl group, 2-methylpyrrol-4-yl group, 2-methyl-pyrrol-5-yl group, 3-methylpyrrol-1-yl group, 3-methyl-pyrrol-2-yl group, 3-methylpyrrol-4-yl group, 3-methylpyrrol-5-yl group, 2-t-butylpyrrol-4-yl group, 3-(2-phenylpropyl)pyrrol-1-yl group, 2-methyl-1-indolyl group, 4-methyl-1-indolyl group, 2-methyl-3-indolyl group, 4-methyl-3-indolyl group, 2-t-butyl-1-indolyl group, 4-t-butyl-1-indolyl group, 2-t-butyl-3-indolyl group, 4-t-butyl-3-indolyl group, and the like.

[0029]

The alkenyl group includes vinyl group, allyl group, 1-butenyl group, 2-butenyl group, 3-butenyl 1,3-butanedienyl group, 1-methylvinyl group, styryl group, 2,2-diphenylvinyl group, 2,2-ditolylvinyl group, 1,2-ditolylvinyl group, 1-methylallyl group, 1-dimethylallyl group, 2-methylallyl group, 1-phenylallyl 2-phenylallyl group, 3-phenylallyl group, 3,3-diphenylallyl group, 1,2-dimethylallyl group, 1-phenyl-1-butenyl group, 3-phenyl-1-butenyl group, and the like.

[0030]

The cycloalkyl group includes cyclopropyl group, cyclobutyl group, cyclopentyl group, cyclohexyl group, 4-methylcyclohexyl group, and the like.

The arylamino group includes, for example, diphenylamino group, ditolylamino group, dinaphthylamino group and naphthylphenylamino group.

The alkylamino group includes, for example, dimethylamino group, diethylamino group and dihexylamino group.

[0031]

The styryl derivative and the aromatic amine include, for example, compounds represented by the following general formulae [vi] and [vii]:

[Chemical Formula 14]

$$Ar^{5'}$$
  $Ar^{6'}$   $Ar^{7'}$   $g$   $vi$ 

[in the formula, Ar<sup>5'</sup>, Ar<sup>6'</sup> and Ar<sup>7'</sup> each independently represent a substituted or unsubstituted aromatic group having 6 to 40 carbon atoms, at least one of which may have styryl group, and g represents an integer of 1 to 4].

[0032]

[Chemical Formula 15]

$$Ar^{8'} \xrightarrow{\left(\begin{array}{c} N \\ Ar^{9'} \end{array}\right)_h} \left(\begin{array}{c} Ar^{10'} \\ Ar^{11'} \end{array}\right)_i \left(\begin{array}{c} N \\ Ar^{12'} \end{array}\right)_j \left(\begin{array}{c} N \\ Ar^{13'} \end{array}\right)_k} Ar^{14'}$$

[vii]

[In the formula,  $Ar^{8'}$ ,  $Ar^{9'}$ ,  $Ar^{11'}$ ,  $Ar^{13'}$  and  $Ar^{14'}$  each independently represent a substituted or unsubstituted monovalent aromatic group having 6 to 40 carbon atoms,  $Ar^{10'}$  and  $Ar^{12'}$  each independently represent a substituted or unsubstituted divalent aromatic group having 6 to 40 carbon atoms, at least one of the groups represented by  $Ar^{8'}$  to  $Ar^{14'}$  may have styryl group or styrylene group, and h and k each represent an integer of 0 to 2].

[0033]

The aromatic group include groups derived from, for example, benzene, naphthalene, anthracene, phenanthrene, pyrene, coronene, biphenyl, terphenyl, pyrrole, furan, thiophene, benzothiophene, oxadiazoline, diphenylanthracene, indoline, carbazole, pyridine, benzoquinone, fluoranthene and acenaphthofluoranthene.

[0034]

In the following, the dopant which can be added to the light emitting layer in the organic EL device of the present invention will be described. The dopant include dopants for

the blue color and dopants for the yellow to red color, and the like.

The dopant for the blue color is not particularly limited.

Dopants satisfying the above relation (V) are preferable.

[0035]

Specific examples of the dopant for the blue color include styrylamine, styryl compounds substituted with an amine and compounds having a condensed aromatic ring.

The styrylamine and the styryl compound substituted with an amine include, for example, compounds represented by the following general formulae [ix] and [x]. The compound having a condensed aromatic ring includes, for example, compounds represented by general formula [ix] shown also in the following.

[Chemical Formula 16]

$$Ar^{1"}$$
 $Ar^{2"}$ 
 $Ar^{3"}$ 
 $p'$ 

[in the formula,  $Ar^{1''}$ ,  $Ar^{2''}$  and  $Ar^{3''}$  each independently represent a substituted or unsubstituted aromatic group having 6 to 40 carbon atoms, at least one of which has styryl group, and p' represents an integer of 1 to 3].

Specific examples of the aromatic group include the groups described above as the examples of the aromatic group.

[0036]

[Chemical Formula 17]

$$U \longrightarrow Ar^{4"} \xrightarrow{C} C \longrightarrow C \longrightarrow Ar^{5"} \xrightarrow{Q'} V$$

$$(x)$$

[in the formula,  $Ar^{4''}$  and  $Ar^{5''}$  each independently represent an arylene group having 6 to 30 carbon atoms,  $E^1$  and  $E^2$  each independently represent an aryl group or alkyl group having 6 to 30 carbon atoms, a hydrogen atom or a cyano group, q' represents an integer of 1 to 3, and U and/or V represents a substituent including an amino group. It is preferable that the amino group is an arylamino group].

Specific examples of the arylene group include divalent derivatives of the groups described above as the specific examples of the aryl group. Specific examples of the aryl group, the alkyl group and the arylamino group include the groups described above as the examples of the corresponding groups.

[0037]

$$(A)_r-B$$
 [xi]

[In the formula, A represents an alkyl group or alkoxyl group having 1 to 16 carbon atoms, a substituted or unsubstituted aryl group having 6 to 30 carbon atoms, a substituted or

unsubstituted alkylamino group having 6 to 30 carbon atoms or a substituted or unsubstituted arylamino group having 6 to 30 carbon atoms, B represents a condensed aromatic cyclic group having 10 to 40 carbon atoms, and r represents an integer of 1 to 4].

Specific examples of the alkyl group, the alkoxyl group, the aryl group, the alkylamino group and the arylamino group include the groups described above as the examples of the corresponding groups.

Other compounds such as fluoresceine derivatives, perylene derivatives or coumarin derivatives can also be used.

Complex compounds shown in the following are also preferable as the dopant.

[Chemical Formula 18]

[0038]

The dopant for a green to red color is not particularly limited. Compounds having an energy gap Eg smaller than 2.65 eV are preferable. The dopant for a green to red color includes fluorescent compounds having a skeleton structure of fluoranthene or perylene. Preferable examples include

compounds represented by the following general formulae [1] to [18].

[Chemical Formula 19]

## [0039]

## [Chemical Formula 20]

$$X^{1}$$
 $X^{12}$ 
 $X^{11}$ 
 $X^{10}$ 
 $X^{9}$ 
 $X^{8}$ 
 $X^{7}$ 
 $X^{7}$ 
 $X^{10}$ 
 $X^{10}$ 

$$X^{1}$$
 $X^{14}$ 
 $X^{13}$ 
 $X^{12}$ 
 $X^{11}$ 
 $X^{10}$ 
 $X^$ 

$$X^{11}$$
  $X^{10}$   $X^{8}$  [8]  $X^{3}$   $X^{4}$   $X^{5}$ 

$$X^{12}$$
 $X^{11}$ 
 $X^{10}$ 
 $X^{9}$ 
 $X^{8}$ 
 $X^{7}$ 
 $X^{10}$ 
 $X^{1$ 

## [0040]

### [Chemical Formula 21]

$$x^{14}$$
 $x^{13}$ 
 $x^{12}$ 
 $x^{10}$ 
 $x$ 

$$X^{1} \xrightarrow{X^{16}} X^{15} \xrightarrow{X^{14}} X^{13} \xrightarrow{X^{12}} X^{11} \xrightarrow{X^{10}} X^{10}$$

$$X^{2} \xrightarrow{X^{3}} X^{4} \xrightarrow{X^{5}} X^{6} \xrightarrow{X^{7}} X^{8} \xrightarrow{X^{8}} X^{9}$$
[12]

$$X^{1}$$
 $X^{14}$ 
 $X^{13}$ 
 $X^{12}$ 
 $X^{11}$ 
 $X^{10}$ 
 $X^$ 

[0041]

#### [Chemical Formula 22]

$$X^{2}$$
 $X^{1}$ 
 $X^{20}$ 
 $X^{19}$ 
 $X^{18}$ 
 $X^{17}$ 
 $X^{16}$ 
 $X^{15}$ 
 $X^{14}$ 
 $X^{13}$ 
 $X^{12}$ 
 $X^{12}$ 
 $X^{14}$ 
 $X^{15}$ 
 $X^{$ 

$$X^{1}$$
 $X^{2}$ 
 $X^{3}$ 
 $X^{4}$ 
 $X^{5}$ 
 $X^{8}$ 
 $X^{7}$ 
 $X^{10}$ 
 $X^{10}$ 

[In the general formulae [1] and [16], X¹ to X²⁰ each independently represent a hydrogen atom, a substituted or unsubstituted linear, branched or cyclic alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted linear, branched or cyclic alkoxyl group having 1 to 20 carbon atoms, a substituted or unsubstituted aryl group having 6 to 30 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted arylamino group having 6 to 30 carbon atoms, a substituted or unsubstituted or unsubstituted or unsubstituted arylamino

unsubstituted alkylamino group having 1 to 30 carbon atoms, a substituted or unsubstituted arylalkylamino group having 7 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 8 to 30 carbon atoms; adjacent substituents and groups represented by  $X^1$  to  $X^{20}$  may be bonded to each other to form a cyclic structure; and when adjacent substituents are aryl groups, the substituents may be the same with each other].

Specific examples of the alkyl group, the alkoxyl group, the aryl group, the aryloxy group, the arylamino group, the alkylamino group and the alkenyl group include the groups described above as the examples of the corresponding groups. Specific examples of the arylalkylamino group include groups obtained by substituting hydrogen atom in an amino group with the alkyl group and the aryl group which are described above.

[0042]

[Chemical Formula 23]

[In the above general formulae [17] and [18],  $R^{1'''}$  to  $R^{4'''}$  each independently represent a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms or a substituted or unsubstituted aryl group having 6 to 30 carbon atoms, a pair of groups represented by  $R^{1'}$  and  $R^{2'}$  and/or  $R^{3'}$  and  $R^{4'}$  may be bonded to each other through the carbon-carbon bond, -0or -S-;  $R^{5'''}$  to  $R^{16'''}$  each independently represent a hydrogen atom, a substituted or unsubstituted linear, branched or cyclic alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted linear, branched or cyclic alkoxyl group having 1 to 20 carbon atoms, a substituted or unsubstituted aryl group having 6 to 30 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted arylamino group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylamino group having 1 to 30 carbon atoms, a substituted or unsubstituted arylalkylamino group having 7 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 8 to 30 carbon atoms; and adjacent substituents and groups represented by  $R^{5^{\prime\prime\prime}}$  to  $R^{16^{\prime\prime\prime}}$ may be bonded to each other to form a cyclic structure. It is preferable that at least one of the groups represented by  $R^{5'''}$  to  $R^{16'''}$  has an amino group or an alkenyl group].

Specific examples of the alkyl group, the alkoxyl group, the aryl group, the aryloxy group, the arylamino group, the alkylamino group and the alkenyl group include the groups

described above as the examples of the corresponding groups. Specific examples of the arylalkylamino group include groups obtained by substituting hydrogen atom in an amino group with the alkyl group or the aryl group which is described above.

[0043]

As other compounds which can be used as the dopant for a yellow to red color, for example, dicyanomethylenepyran derivatives, dicyanomethylenethiopyran derivatives, fluoresceine derivatives, perylene derivatives and coumarin derivatives which are described in European Patent Application Laid-Open No. 0281381 and used as red or reddish laser dyes are also preferable. Specific examples of the compound include the following compounds:

# [Chemical Formula 24]

(Lumogen F red)

$$(H_5C_2)_2N \longrightarrow C = C \longrightarrow CH_2$$

$$CN \longrightarrow CN$$

(dicyanomethylenepyran)

$$(H_5C_2)_2N$$
 O O

(phenoxazone)

(rubrene)

Lumogen F red: Eg(ETM)=2.0 Ev

rubrene: Eg(ETM)=2.2 eV

and, in addition, other compounds such as:
[Chemical Formula 25]

$$\begin{bmatrix} \\ \\ \\ \\ \\ \end{bmatrix}_3$$

[0044]

In the following, the heterocyclic compound having nitrogen atom which is comprised in the electron transporting layer in the present invention will be described.

It is preferable that the electron transporting layer in the organic EL device of the present invention comprises a heterocyclic derivative having nitrogen atom represented by the following general formula (1):

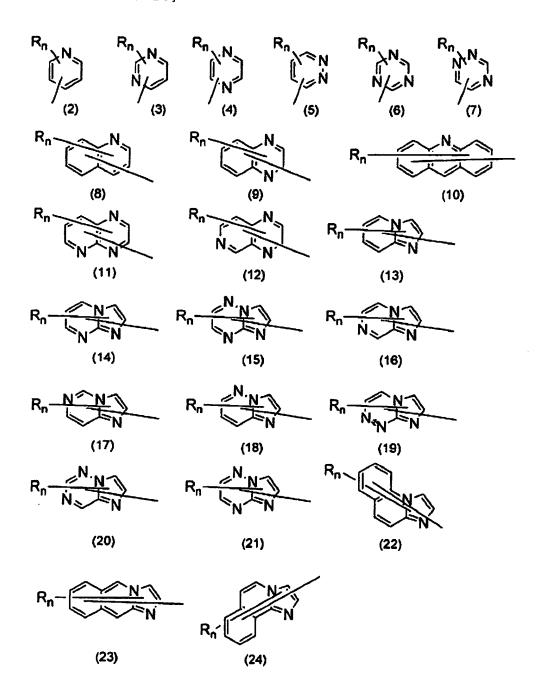
$$HAr-L-Ar^1-Ar^2$$
 (1)

(in the formula, HAr represents a substituted or unsubstituted heterocyclic group having nitrogen atom, Ar<sup>1</sup> represents a substituted or unsubstituted divalent aromatic hydrocarbon group having 6 to 40 carbon atoms, Ar<sup>2</sup> represents a substituted or unsubstituted aryl group having 6 to 40 carbon atoms or a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms, and L represents a single bond or a substituted or unsubstituted arylene group).

[0045]

It is preferable that, in the general formula (1), HAr represents a heterocyclic group having nitrogen atom represented by one of the following formulae (2) to (24) and the following formula (A):

### [Chemical Formula 26]



(in the formula, R represents a substituted or unsubstituted aryl group having 6 to 40 carbon atoms, a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms or a substituted or unsubstituted alkoxyl group having 1 to 20 carbon atoms, n represents an integer of 0 to 5 and, when n represents an integer of 2 or greater, plural R may represent the same group or different groups, and plural groups represented by R may be bonded to each other to form a cyclic structure).

[0046]

[Chemical Formula 27]

$$\begin{array}{c|c} & & \\ \hline & & \\ Z & & \\ \hline \end{array} \begin{array}{c} R' \\ \end{array} \begin{array}{c} \\ X \end{array} \qquad (A)$$

(in the formula, plural  $R^1$  each independently represent a hydrogen atom, a halogen atom, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted aryl group having 6 to 40 carbon atoms, a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms or a group forming a condensed aromatic ring, Z represents an oxygen atom, a sulfur atom or a group represented by NR', (R' representing the same atom or group as that represented by  $R^1$ ), and x represents an integer of 2 to 8);

or a substituted or unsubstituted carbazolyl group.

Specific examples of the aryl group, the heteroaryl group, the alkyl group and the alkoxyl group include the groups described above as the examples of the corresponding groups.

Examples of the substituent to carbazolyl group include the groups described above as the examples of the groups represented by  $R^\prime$ .

[0047]

It is more preferable that the HAr represents a group expressed by one of the following formulae (25) to (44):

### [Chemical Formula 28]

[0048]

It is preferable that, in general formula (1), the L represents a group represented by the following formula (45) or (46):

[Chemical Formula 29]

$$\begin{array}{c|cccc}
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(in the formula,  $R^2$  represents a substituted or unsubstituted aryl group having 6 to 40 carbon atoms, a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms or a substituted or unsubstituted alkoxyl group having 1 to 20 carbon atoms, m represents an integer of 0 to 4 and, when m represents an integer of 2 or greater, plural  $R^2$  may represent the same group or different groups, and plural groups represented by  $R^2$  may be bonded to each other to form a cyclic structure).

Specific examples of the aryl group, the heteroaryl group, the alkyl group and the alkoxyl group include the groups described above as the examples of the corresponding groups.

[0049]

It is preferable that, in general formula (1), the  ${\rm Ar}^2$  represents a group represented by one of the following formulae

(47) to (53):

[Chemical Formula 30]

$$R^{3}p$$
 $R^{3}p$ 
 $R^{3}p$ 

(in the formula,  $R^3$  represents a substituted or unsubstituted aryl group having 6 to 40 carbon atoms, a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms or a substituted or unsubstituted alkoxyl group having 1 to 20 carbon atoms, p represents an integer of 0 to 9, q represents an integer of 0 to 5 and, when p or p+q represents an integer of 2 or greater, plural  $R^3$  may represent the same group or different groups, and plural groups represented by  $R^3$  may be bonded to each other to form a cyclic structure).

Specific examples of the aryl group, the heteroaryl group, the alkyl group and the alkoxyl group include the groups described above as the examples of the corresponding groups.

[0050]

It is preferable that, in general formula (1), the  $Ar^1$  represents a group represented by the following formula (54) or (55):

[Chemical Formula 31]

$$R^{5}$$
 $R^{6}$ 
 $R^{7}$ 
 $R^{8}$ 
 $R^{9}$ 
 $R^{10}$ 
 $R^{12}$ 
 $R^{17}$ 
 $R^{17}$ 
 $R^{18}$ 
 $R^{16}$ 
 $R^{15}$ 
 $R^{15}$ 
 $R^{15}$ 

(in the formula, R<sup>4</sup> to R<sup>17</sup> each independently represent a hydrogen atom, a halogen atom, a substituted or unsubstituted aryl group having 6 to 40 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 40 carbon atoms, a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms or a substituted or unsubstituted alkoxyl group having 1 to 20 carbon atoms, and Ar<sup>3</sup> and Ar<sup>4</sup> each represent a substituted or unsubstituted aryl group having 6 to 40 carbon atoms or a substituted or unsubstituted heteroaryl group having 3 to 40 carbon atoms.

Specific examples of the aryl group, the heteroaryl group, the alkyl group and the alkoxyl group include the groups described above as the examples of the corresponding groups. Examples of the halogen atom include fluorine atom, chlorine

atom, bromine atom, iodine atom, and the like.

[0051]

Specific examples of the heterocyclic derivative having silicon atom comprised in the electron transporting layer in the present invention include the compounds described in Japanese Patent Application Laid-Open No. Heisei 09(1997)-194487.

Examples of the above compound include silacyclopentadiene derivatives represented by the following general formula:

[Chemical Formula 32]

$$R_3$$
 $R_4$ 
 $X$ 
 $Si$ 
 $Y$ 

[in the formula, X and Y each independently represents a saturated or unsaturated hydrocarbon group having 1 to 6 carbon atoms, an alkoxyl group, an alkenyloxy group, an alkynyloxy group, a substituted or unsubstituted aryl group, a substituted or unsubstituted hetero ring or a structure formed by bonding the groups represented by X and Y to form a saturated or unsaturated ring; and  $R_1$  to  $R_4$  each independently represent a hydrogen atom, a halogen atom, a substituted or unsubstituted alkyl group having 1 to 6 carbon atoms, an alkoxy group, an

aryloxy group, a perfluoroalkyl group, a perfluoroalkoxy group, an amino group, an alkylcarbonyl group, an arylcarbonyl group, an alkoxycarbonyl group, an aryloxycarbonyl group, an azo group, an alkylcarbonyloxy group, an arylcarbonyloxy group, alkoxycarbonyloxy group, an aryloxycarbonyloxy group, a sulfinyl group, a sulfonyl group, a sulfanyl group, a silyl group, a carbamoyl group, an aryl group, a heterocyclic group, an alkenyl group, an alkynyl group, a nitro group, a formyl group, a nitroso group, a formyloxy group, an isocyano group, a cyanate group, an isocyanate group, a thiocyanate group, an isothiocyanate group, a cyano group or, when the groups are adjacent to each other, a structure formed by condensation of substituted or unsubstituted rings (however, when  $R_1$  and  $R_4$ represent phenyl groups, X and Y do not represent alkyl groups or phenyl groups. When R1 and R4 represent thienyl groups, X and Y represent monovalent hydrocarbon groups, and R2 and R3 represent alkyl groups, aryl groups, alkenyl groups or a structure formed by bonding of the groups represented by R2 and R3 to form a ring in which the structures derived from the groups represented by  $R_2$  and  $R_3$  do not satisfy the condition of the aliphatic group simultaneously. When  $R_1$  and  $R_4$  represent silyl groups,  $R_2$ ,  $R_3$ , X and Y each independently do not represent a monovalent hydrocarbon group having 1 to 6 carbon atoms or a hydrogen atom. When  $R_1$  and  $R_2$  represent in combination a structure formed by condensation of the benzene rings, X and

Y do not represent alkyl groups or phenyl groups)].

[0052]

In the construction of the organic EL device of the present invention, the hole injecting layer and the electron injecting layer are not always necessary. However, the device comprising these layers has an advantage in that the light emitting property is improved. A hole transporting layer (a hole injecting layer), a light emitting layer and an electron injecting layer may be disposed between a pair of electrodes in a mixed form. Mixed layers may be formed using a binder such as a macromolecular compound so that the constituting components can be present with stability.

The organic EL device of the present invention will be described with reference to an embodiment having a construction of an anode/a hole transporting layer/a light emitting layer/an electron transporting layer/a cathode. It is preferable that the organic EL device of the present invention is supported with a substrate. The substrate is not particularly limited, and a substrate conventionally used for organic EL devices made such as of glass, a transparent plastic material and quartz can be used.

[0053]

As the anode in the organic EL device, an anode using a metal, an alloy, an electrically conductive compound or a mixture of these substances, which has a great work function

(4 eV or greater), as the electrode material is preferably used. Specific examples of the electrode material include metals such as Au and conductive transparent materials such as CuI, ITO, SnO2 and ZnO. The anode can be prepared by forming a thin film of the electrode material described above in accordance with a process such as the vapor deposition process and the sputtering process. When the light emission is obtained through the anode, it is preferable that the transmittance is greater than 10%. It is also preferable that the sheet resistivity as the electrode is several hundred  $\Omega/\Box$  or smaller. The thickness of the anode is, in general, selected in the range of 10 nm to 1  $\mu$ m and preferably in the range of 10 to 200 nm although the preferable range depends on the used material.

[0054]

As the cathode, a cathode using a metal, an alloy, an electrically conductive compound or a mixture of these substances, which has a small work function (4 eV or smaller), as the electrode material is used. Specific examples of the electrode material include sodium, sodium-potassium alloys, magnesium, magnesium-silver alloys, lithium, magnesium/copper mixtures, magnesium-indium alloys, Al/Al<sub>2</sub>O<sub>3</sub>, indium and aluminum-lithium alloys. The cathode can be prepared by forming a thin film of the electrode material described above in accordance with a process such as the vapor deposition process and the sputtering process. It is

preferable that the sheet resistivity as the electrode is several hundred  $\Omega/\Box$  or smaller. The film thickness is selected, in general, in the range of 10 to 500 nm and preferably in the range of 50 to 200 nm. For transmitting light emission, it is advantageous that one of the anode and the cathode of the organic EL device is transparent or translucent so that the efficiency of light emission is increased.

[0055]

As the light emitting material constituting the light emitting layer in the organic EL device of the present invention, a host material as described above is used. As the host material, in addition to the dopants as described above, a light emitting material selected as desired from heretofore known light emitting materials can be used. As the light emitting material, for example, polycyclic condensed aromatic whitening compounds, fluorescent agents such as benzoxazole-based agents, benzothiazole-based agents and benzimidazole-based agents, metal chelated oxanoid compounds and distyrylbenzene-based compounds, which provide excellent thin film-forming properties, can be used. Examples of the polycyclic condensed aromatic compound include condensed cyclic light emitting substances having a skeleton structure derived from anthracene, naphthalene, phenanthrene, pyrene, chrysene or perylene, and other condensed cyclic light emitting substances having about 8 condensed rings. Specifically,

1,1,4,4-tetraphenyl-1,3-butadiene

and

4,4'-(2,2-diphenyl-vinyl)biphenyl can be used. The light emitting layer may be constituted with a single layer comprising one or at least two of these light emitting materials or may be a laminate with light emitting layers comprising compounds other than those of the light emitting layers described above.

[0056]

The hole transporting layer (the hole injecting layer) in the organic EL device of the present invention comprises a hole transporting compound and exhibits the function of transporting the hole injected from the anode to the light emitting layer. Due to the hole injecting layer disposed between the anode and the light emitting layer, a greater amount of holes can be injected into the light emitting layer under application of a lower electric field. Moreover, the electrons injected into the light emitting layer from the cathode or the electron injecting layer are accumulated in the interfacial region in the light emitting layer due to the electron barrier existing at the interface between the light emitting layer and the hole transporting layer (the hole injecting layer), so that a device exhibiting a more excellent light emitting property such as an increased efficiency of light emission can be obtained. The hole transporting compound used for the hole transporting layer (the hole

injecting layer) is a compound which is disposed between the two electrodes to which an electric field is applied and can suitably transport holes to the light emitting layer when the holes are injected from the anode. For example, compounds having a mobility of holes of at least  $10^{-6}$  cm<sup>2</sup>/V·sec under application of an electric field of  $10^4$  to  $10^6$  V/cm are preferable. The hole transporting compound is particularly limited as long as the compound has the above preferable property. The hole transporting compound can be selected as desired from compounds conventionally used as the material for charge injection and transportation of holes in photoconductive materials and heretofore known compounds used for the hole transporting layer (the hole injecting layer) of organic EL devices.

[0057]

The hole transporting compound includes, for example, copper phthalocyanine, N,N,N',N'-tetraphenyl-4,4'-diaminophenyl, N,N'-diphenyl-N,N'-di(3-methylphenyl)-4,4'-diaminobiphenyl (TPDA), 2,2-bis(4-di-p- tolylaminophenyl)propane, 1,1-bis(4-di-p-tolylaminophenyl)cyclohexane and N,N,N',N'-tetra-p-tolyl-4,4'-diaminobiphenyl. Crystals and amorphous materials of inorganic semiconductors such as Si, SiC and CdS can also be used. The hole injecting layer may be constituted with a single layer comprising one or at least

two of these hole injecting materials or may be a laminate with a hole injecting layer comprising compounds other than those of the hole injection layer described above.

[0058]

As a preferable embodiment of the organic EL device of the present invention, the organic EL device may comprise a reducing dopant in the region of transporting electrons or in the interface region of the cathode and an organic compound layer. In the present invention, an organic EL device in which the compound of the present invention comprises a reducing dopant is preferable. The reducing dopant is defined as a substance which can reduce a compound having the electron transporting property. Therefore, various compounds can be used as long as the compounds have a prescribed reducing property. For example, at least one substance selected from the group consisting of alkali metals, alkaline earth metals, rare earth metals, oxides of alkali metals, halides of alkali metals, oxides of alkaline earth metals, halides of alkaline earth metals, oxides of rare earth metals, halides of rare earth metals, organic complexes of alkali metals, organic complexes of alkaline earth metals and organic complexes of rare earth metals is preferable.

[0059]

As the reducing dopant, substances having a work function of 2.9 eV or smaller are preferable. More specifically, at

least one alkali metal selected from the group consisting of Na (the work function: 2.36 eV), K (the work function: 2.28 eV), Rb (the work function: 2.16 eV) and Cs (the work function: 1.95 eV) and at least one alkaline earth metal selected from the group consisting of Ca (the work function: 2.9 eV), Sr (the work function: 2.0 to 2.5 eV) and Ba (the work function: 2.52 Among the above substances, more eV) are preferable. preferable reducing dopants is at least one alkali metal selected from the group consisting of K, Rb and Cs, and further preferable dopants are Rb and Cs, and the most preferable These alkali metals have great reducing dopants is Cs. reducing ability, and the luminance of the emitted light and the lifetime of the organic EL device can be increased by addition of a relatively small amount of the alkali metal into the electron injecting zone. As the reducing dopant having a work function of 2.9 eV or smaller, combinations of two or more of the alkali metals are also preferable, and combinations having Cs as a component such as the combinations of Cs and Na, Cs and K, Cs and Rb or Cs, Na and K are more preferable. The reducing ability can be efficiently exhibited by the combination having Cs as a component. The luminance of emitted light and the lifetime of the organic EL device can be increased by the addition of the combination into the electron injecting zone. The same effects can be exhibited by using, other than the alkali metals, at least one metal compound selected from

the group consisting of chalcogenides of alkali metals, chalcogenides of alkaline earth metals, halides of alkali metals and halides of alkaline earth metals. The same effects can also be exhibited by using organic complexes of alkali metals and organic complexes of alkaline earth metals.

[0060]

The organic EL device of the present invention may further comprise an electron injecting layer which is constituted with an insulating material or a semiconductor and disposed between the cathode and the organic layer. Ву disposing the electron injecting layer, leak of the electric current can be effectively prevented, and the electron injecting property can be improved. As the insulating material, at least one metal compound selected from the group consisting of chalcogenides of alkali metals, chalcogenides of alkaline earth metals, halides of alkali metals and halides of alkaline earth metals is preferable. It is preferable that the electron injecting layer is constituted with the above metal compound since the electron injecting property can be further improved. Preferable examples of the chalcogenide of an alkali metal include, specifically, Li<sub>2</sub>O, LiO, Na<sub>2</sub>S, Na<sub>2</sub>Se Preferable examples of the chalcogenide of an and NaO. alkaline earth metal include CaO, BaO, SrO, BeO, BaS and CaSe. Preferable examples of the halide of an alkali metal include LiF, NaF, KF, LiCl, KCl and NaCl. Preferable examples of the

halide of an alkaline earth metal include fluorides such as  $\text{CaF}_2$ ,  $\text{BaF}_2$ ,  $\text{SrF}_2$ ,  $\text{MgF}_2$  and  $\text{BeF}_2$  and halides other than the fluorides.

[0061]

Examples of the semiconductor constituting the electron transporting layer include oxides, nitrides or oxynitrides comprising at least one element selected from Ba, Ca, Sr, Yb, Al, Ga, In, Li, Na, Cd, Mg, Si, Ta, Sb and Zn used singly or in combination of two or more. It is preferable that the inorganic compound constituting the electron injecting layer forms a finely crystalline or amorphous insulating thin film. When the electron injecting layer is constituted with the inorganic compound described above, a more uniform thin film can be formed, and defects of pixels such as dark spots can be decreased. Examples of the inorganic compound include chalcogenides of alkali metals, chalcogenides of alkaline earth metals, halides of alkali metals and halides of alkaline earth metals which are described above.

[0062]

The process for preparing the organic EL device of the present invention will be described in the following. As a preferable embodiment, a process for preparing an organic EL device having a construction of an anode/a hole transporting layer/a light emitting layer/an electron transporting layer/a cathode, which is described above, will be described in the

following. On a suitable substrate, a thin film made of a prescribed material for the electrode, for example, a material for the anode is formed in accordance with the vapor deposition process or the sputtering process so that the thickness of the formed thin film is 1 µm or smaller and preferably in the range of 10 to 200 nm to form an anode. On the formed anode, a hole transporting layer, plural emitting layers and an electron transporting layer as the constituting elements of the EL device are formed by successively laminating thin layers comprising the respective constituting materials. process for the formation of the thin layers, the spin coating process, the casting process or the vapor deposition process can be used as described above. The vacuum vapor deposition process is preferable since a uniform film can be easily obtained and the possibility of formation of pin holes is small. When the thin layers are formed in accordance with the vacuum vapor deposition process, in general, it is preferable that the conditions are suitably selected in the following ranges: the temperature of heating the port: 50 to  $400^{\circ}$ C; the degree of vacuum:  $10^{-6}$  to  $10^{-3}$  Pa; the rate of deposition: 0.01 to 50 nm/second; the temperature of the substrate: -50 to  $300^{\circ}$ ; and the thickness of the film: 5 nm to  $5 \mu m$ ; although the conditions of the vacuum vapor deposition are different depending on the type of the used compound and the crystal structure and the aggregation structure of the molecular accumulation film to

be formed. After these layers are formed, a thin layer having a thickness of 1  $\mu m$  or smaller and preferably in the range of 50 to 200 nm and comprising the substance for the cathode is formed in accordance with the vapor deposition process or the sputtering process to form the cathode, whereby the desired organic EL device can be obtained. The organic EL device may be prepared in the order reverse to that described above, i.e., in the order of the cathode, the electron transporting layer, the light emitting layer, the hole transporting layer and the anode.

[0063]

[Examples]

The present invention will be described more specifically with reference to examples in the following. However, the present invention is not limited to the examples.

The energy gap and the ionization potential of a compound were measured in accordance with the following methods.

## (1) Measurement of the energy gap (Eg)

The energy gap of the excited singlet was measured. That is, an absorption spectrum of a toluene solution (10<sup>-5</sup> mole/liter) of a sample was obtained using an ultraviolet visible absorptiometer manufactured by HITACHI SEISAKUSHO Co., Ltd. A tangent was drawn to the increasing curve of the spectrum at the side of the long wavelength, and the wavelength at the intersection of the tangent with the abscissa (the end

of the absorption) was obtained. The obtained wavelength was converted into the energy value.

## (2) Measurement of the ionization potential (Ip)

A curve showing the change in the discharge of photoelectrons with the photon energy of irradiation was obtained with respect to a compound for the measurement using an atmospheric photoelectron spectrometer AC-1 manufactured by RIKEN KEIKI Co., Ltd., and the threshold value of the discharge of photoelectrons was obtained from the obtained curve in accordance with the extrapolation method.

[0064]

Example 1 (Preparation of an organic EL device of the laminate type having a light emitting layer emitting blue light/a light emitting layer emitting yellow light)

A glass substrate of 25 mm  $\times$  75 mm  $\times$  1.1 mm thickness having an ITO transparent electrode (manufactured by GEOMATEC Company) was cleaned by application of ultrasonic wave in isopropyl alcohol for 5 minutes and then by exposure to ozone generated by ultraviolet light for 30 minutes. The cleaned glass substrate having a transparent electrode line was attached to a substrate holder of a vacuum vapor deposition apparatus. On the surface of the cleaned substrate at the side having the transparent electrode line, a film of N,N'-bis(N,N'-diphenyl-4-aminophenyl)-

N, N-diphenyl-4,4'-diamino-1,1'-biphenyl (abbreviated as

"TPD232 film", hereinafter) having a thickness of 60 nm was formed in a manner such that the formed film covered the transparent electrode. The TPD232 film worked as the hole film of injecting layer. On the TPD232 film, N, N, N', N'-tetra(4-biphenyl) diaminobiphenyl (abbreviated as "TBTB film", hereinafter; Ip=5.5 eV; Eg=3.1 eV) having a thickness of 20 nm was formed. The TBTB film worked as the hole transporting layer. On the TBTB film, 4',4"-bis(2,2-diphenylvinyl)-9,10-diphenylanthracene (DPVDPAN) (Ip=5.7 eV; Eg=3.0 eV) as the styryl derivative and Compound B1 shown in the following (the peak of light emission: 470 nm; Ip=5.5 eV; Eq=2.8 eV) as the dopant compound were vapor deposited simultaneously to form a film having a thickness of 10 nm with the ratio of the film thickness of 40:2. The formed film was used as a first light emitting layer (the light emitting layer emitting blue light). Then, DPVDPAN (Ip=5.7 eV; Eq=3.0 eV) as the styryl derivative and Compound R1 shown in the following (the peak of light emission: 560 nm; Ip=5.6 eV; Eg=2.6 eV) as the dopant compound were vapor deposited simultaneously to form a film having a thickness of 30 nm with the film thickness ratio of 40:2. The formed film was used as a second light emitting layer (the light emitting layer emitting yellow light). On the formed film, a film of ETM20 (Ip=5.7 eV; Eq=3.0 eV) having a thickness of 20 nm was formed as an electron transporting layer. Then, LiF was vapor

deposited to form a film having a thickness of 1 nm in accordance with the resistance heating method, whereby an electron injecting cathode was formed. On the formed film, metallic aluminum was vapor deposited to form a film having a thickness of 150 nm, whereby a metal cathode was formed. Thus, an organic EL device emitting white light was prepared.

The obtained organic EL device was examined by passing electric current. Emission of white light having a luminance of 1,000 nit was obtained at a voltage of 5.0 V and a current density of 11.0 mA/cm². The chromaticity coordinates were (0.33, 0.33), and the efficiency of light emission was 9.1 cd/A and 5.7 lumen/W. The luminance was measured after driving for 1,500 hours at the room temperature under the condition of the initial luminance of 1,000 nit and was found to be 700 nit.

[0065]

## [Chemical Formula 33]

B1 R1

ETM20

[0066]

Example 2 (Preparation of an organic EL device of the laminate type having a light emitting layer emitting blue light/a light emitting layer emitting yellow light/a light emitting layer emitting blue light)

An organic EL device emitting white light was prepared in accordance with the same procedures as those conducted in Example 1 except that the light emitting layers were prepared as described in the following.

That is, on the TBTB film, DPVDPAN (Ip=5.7 eV; Eg=3.0 eV) as the styryl derivative and Compound B1 shown above (the

peak of light emission: 470 nm; Ip=5.5 eV; Eg=2.8 eV) as the dopant compound were vapor deposited simultaneously to form a film having a thickness of 10 nm with the film thickness ratio of 40:2. The formed film was used as a first light emitting layer (the light emitting layer emitting blue light). Then, DPVDPAN (Ip=5.7 eV; Eg=3.0 eV) as the styryl derivative and Compound R1 shown above (the peak of light emission: 560 nm; Ip=5.6 eV; Eg=2.6 eV) as the dopant compound were vapor deposited simultaneously to form a film having a thickness of 10 nm with the film thickness ratio of 40:2. The formed film was used as a second light emitting layer (the light emitting layer emitting yellow light). Thereafter, DPVDPAN (Ip=5.7 eV; Eq=3.0 eV) as the styryl derivative and Compound B1 shown above (the peak of light emission: 470 nm; Ip=5.5 eV; Eg=2.8 eV) as the dopant compound were vapor deposited simultaneously to form a film having a thickness of 20 nm with the film thickness ratio of 40:2. The formed film was used as a third light emitting layer (the light emitting layer emitting blue light).

The obtained organic EL device was examined by passing electric current. Emission of white light having a luminance of 1,000 nit was obtained at a voltage of 4.5 V and a current density of 10.0 mA/cm<sup>2</sup>. The chromaticity coordinates were (0.28, 0.35), and the efficiency of light emission was 10.0 cd/A and 7.0 lumen/W. The luminance was measured after driving for 1,500 hours at the room temperature under the condition

of the initial luminance of 1,000 nit and was found to be 600 nit.

[0067]

C

Example 3 (Preparation of an organic EL device of the laminate type having a light emitting layer emitting yellow light/a light emitting layer emitting blue light/a light emitting layer emitting yellow light)

An organic EL device emitting white light was prepared in accordance with the same procedures as those conducted in Example 1 except that the light emitting layers were prepared as described in the following.

That is, on the TBTB film, DPVDPAN (Ip=5.7 eV; Eg=3.0 eV) as the styryl derivative and Compound R1 shown above (the peak of light emission: 560 nm; Ip=5.6 eV; Eg=2.6 eV) as the dopant compound were vapor deposited simultaneously to form a film having a thickness of 10 nm with the film thickness ratio of 40:2. The formed film was used as a first light emitting layer (the light emitting layer emitting yellow light). Then, DPVDPAN (Ip=5.7 eV; Eg=3.0 eV) as the styryl derivative and Compound B1 shown above (the peak of light emission: 470 nm; Ip=5.5 eV; Eg=2.8 eV) as the dopant compound were vapor deposited simultaneously to form a film having a thickness of 10 nm with the film thickness ratio of 40:2. The formed film was used as a second light emitting layer (the light emitting layer emitting blue light). Thereafter, DPVDPAN (Ip=5.7 eV;

Eg=3.0 eV) as the styryl derivative and Compound R1 shown above (Ip=5.6 eV; Eg=2.6 eV) as the dopant compound were vapor deposited simultaneously to form a film having a thickness of 20 nm with the film thickness ratio of 40:2. The formed film was used as a third light emitting layer (the light emitting layer emitting yellow light).

The obtained organic EL device was examined by passing electric current. Emission of white light having a luminance of 1,000 nit was obtained at a voltage of 5.3 V and a current density of 11.0 mA/cm<sup>2</sup>. The chromaticity coordinates were (0.32, 0.35), and the efficiency of light emission was 9.1 cd/A and 5.4 lumen/W. The luminance was measured after driving for 1,500 hours at the room temperature under the condition of the initial luminance of 1,000 nit and was found to be 800 nit.

[8800]

Comparative Example 1 (Preparation of an organic EL device of the laminate type having a light emitting layer emitting blue light/a light emitting layer emitting green to red light)

A glass substrate (manufactured by GEOMATEC Company) of  $25 \text{ mm} \times 75 \text{ mm} \times 1.1 \text{ mm}$  thickness having an ITO transparent electrode was cleaned by application of ultrasonic wave in isopropyl alcohol for 5 minutes and then by exposure to ozone generated by ultraviolet light for 30 minutes. The cleaned glass substrate having the transparent electrode line was attached to a substrate holder of a vacuum vapor deposition

apparatus. On the surface of the cleaned substrate at the side having a transparent electrode line, a TPD232 film having a thickness of 60 nm was formed in a manner such that the formed The TPD232 film film covered the transparent electrode. worked as the hole injecting layer. On the TPD232 film, a film of 4,4'-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (abbreviated as "NPD film", hereinafter) having a thickness of 20 nm was formed. The NPD film worked as the hole transporting layer. On the formed NPD film, DPVPDAN (Ip=5.7 eV; Eq=3.0 eV) as the styryl derivative and Compound B1 shown above (Ip=5.5 eV; Eq=2.8 eV) were further vapor deposited to form a film having a thickness of 20 nm with the film thickness ratio of 20:1.5. The formed film was used as the light emitting layer emitting blue light. Then, DPVPDAN as the styryl derivative and Compound R1 shown above were vapor deposited to form a film having a thickness of 20 nm with the film thickness ratio of 20:1. The formed film was used as the light emitting layer emitting green to red light. On the formed film, a film of t-BuPBD shown in the following (Ip=6.1 eV; Eg=3.1 eV) having a thickness of 20 nm was formed as an electron transporting layer. Then, LiF (the source of lithium: manufactured by SAES GETTERS Company) was vapor deposited to form a film having a thickness of 1 nm. On the formed film, metallic aluminum was vapor deposited to form a film having a thickness of 150 nm, whereby a metal cathode was formed. Thus, an organic EL device

was prepared.

The obtained organic EL device was examined by passing electric current. Emission of light having a luminance of 110 nit was obtained at a voltage of 7 V and a current density of 2.0 mA/cm². The chromaticity coordinates were (0.25, 0.33), and the efficiency of light emission was 5.5 cd/A and 2.5 lumen/W. The luminance was measured after driving for 1,500 hours at the room temperature under the condition of the initial luminance of 1,000 nit and was found to be 300 nit.

[Chemical Formula 34]

t-BuPBD (t-Bu represents t-butyl group.)

[0069]

Ç,

[Advantage of the Invention]

The organic EL device emitting white light of the present invention provides a great efficiency of light emission under a low voltage, has a long lifetime and exhibits no change in the chromaticity. Therefore, the organic EL device emitting white light of the present invention is advantageous in the practical applications and very useful as the light source, etc. for various types of electronic instruments.

[Brief Description of the Drawings]

[Fig. 1] Fig. 1 is a diagram showing an embodiment of

the construction of the organic electroluminescence device emitting white light of the present invention.

[Fig. 2] Fig. 2 is a diagram showing the area defining the white light in the CIE chromaticity coordinates.

[Designation of Document] Abstract
[Abstract]

[Problem] An organic electroluminescence device emitting white light providing a great efficiency of light emission under a low voltage, having a long lifetime and exhibiting no change in the chromaticity, is provided.

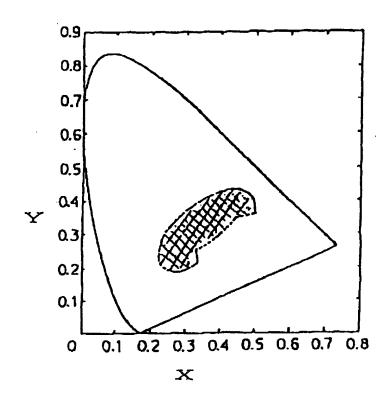
[Means for Resolution] An organic electroluminescence device emitting white light including a pair of electrodes, at least two light emitting layers and an electron transporting layer including a heterocyclic derivative having nitrogen atom or a heterocyclic derivative having silicon atom, the light emitting layers and the electron transporting layer being disposed between the pair of electrodes, wherein the energy gap of a host material contained in the light emitting layer is defined in a specific range, the energy gap of the heterocyclic derivative having nitrogen atom or heterocyclic derivative having silicon atom contained in the electron transporting layer is defined in a specific range, and the ionization potential of the heterocyclic derivative having nitrogen atom or the heterocyclic derivative having silicon atom in the electron transporting layer and the ionization potential of the host material in the light emitting layer adjacent to the electron transporting layer satisfy a specific relation.

[Selected Drawing] None

[Designation of Document] DRAWING [FIG. 1]

CATHODE
(ELECTRON INJECTING LAYER)
ELECTRON TRANSPORTING LAYER
(COMPOUND HAVING NITROGEN ATOM)
THE FIRST LIGHT EMITTING LAYER
THE SECOND LIGHT EMITTING LAYER
THE i-TH LIGHT EMITTING LAYER
THE n-TH LIGHT EMITTING LAYER
(HOLE TRANSPORTING LAYER)
(HOLE INJECTING LAYER)
ANODE

[FIG. 2]



AREA OF THE WHITE LIGHT IN THE CIE COORDINATES